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Zero-temperature Phase Diagram For Strongly-Correlated Nanochains

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Abstract

Recently there has been a resurgence of intense experimental and theoretical interest on the Kondo physics of nanoscopic and mesoscopic systems due to the possibility of making experiments in extremely small samples. We have carried out exact diagonalization calculations to study the effect of the energy spacing Δ of the conduction band on the ground-state properties of a dense Anderson model nanochain. The calculations reveal for the first time that the energy spacing tunes the interplay between the Kondo and RKKY interactions, giving rise to a zero-temperature Δ versus hybridization phase diagram with regions of prevailing Kondo or RKKY correlations, separated by a *free spins* regime. This interplay may be relevant to experimental realizations of small rings or quantum dots with tunable magnetic properties.

The possibility of making experiments in extremely small samples has lead to a resurgence of both experimental and theoretical interest of the physics of the interaction of magnetic impurities in nanoscopic and mesoscopic non-magnetic metallic systems. A few examples include quantum dots[1], quantum boxes[2] and quantum corrals[3]. Recent scanning tunneling microscope(STM) experiments[4] studied the interaction of magnetic impurities with the electrons of a single-walled nanotube confined in one dimension. Interestingly, in addition to the bulk Kondo resonance new subpeaks were found in shortened carbon nanotubes, separated by about the average energy spacing, Δ, in the nanotube. The relevance of small strongly correlated systems to quantum computation requires understanding how the infinite-size properties become modified at the nanoscale, due to the finite energy spacing Δ in the conduction band[2,5,6,7,8]. For such small systems, controlling T_K upon varying Δ is acquiring increasing importance since it allows to tune the cluster magnetic behavior and to encode quantum information. While the effect of Δ on the single-impurity Anderson or Kondo model has received considerable theoretical[2,5,6,7,8] and experimental[4] attention recently, its role on *dense* impurity clusters remains an unexplored area thus far. The low-temperature behavior of a nanosized heavy-electron system was recently studied within the mean-field approximation[9]. A central question is what is the effect of Δ on the interplay between the Kondo effect and the RKKY interaction. The first interaction being responsible for the quenching of the local f-moment (LM) via the screening of the conduction electrons, whereas the latter being responsible for magnetic ordering.

In this work we present exact diagonalization calculations[10,11] for f-electron nanochains using periodic boundary conditions to study the effect of (1) energy spacing, (2) f-electron conduction-electron hybridization and (3) the parity of number of conduction electrons on the interplay between the Kondo and RKKY interactions. While the cluster properties depend on cluster geometry and size[12], the present calculations treat exactly the Kondo and RKKY interactions. Our results show that tuning Δ and the *parity* of the total number of electrons can drive the nanocluster from the Kondo to the RKKY regime, giving rise to a zero-temperature energy spacing versus hybridization phase diagram which is rich in structure.

We consider the half-filled ($N_{el} = 2N$) periodic Anderson Hamiltonian for N=6 sites

arranged in a ring

$$H = t \sum_{<ij>\sigma} c^{\dagger}_{i\sigma} c_{j\sigma} + \sum_{i\sigma} \epsilon^{i}_{f} f^{\dagger}_{i\sigma} f_{i\sigma} + \sum_{i} U_i f^{\dagger}_{i+} f_{i+} f^{\dagger}_{i-} f_{i-} + \sum_{i\sigma} V(f^{\dagger}_{i\sigma} c_{i\sigma} + c^{\dagger}_{i\sigma} f_{i\sigma}). \quad (1)$$

Here, t is the nearest-neighbor hopping matrix element for the conduction electrons, $c^{+}_{i,\sigma}(c_{i,\sigma})$ and $f^{+}_{i,\sigma}(f_{i,\sigma})$ create (annihilate) Wannier electrons in c- and f- like orbitals on site i with spin σ, respectively. E_f is the energy levels of the bare localized orbital, V is the on-site hybridization matrix element between the f and conduction orbitals, and U is the on-site Coulomb repulsion of the f electrons. In this paper we consider a simple tight-binding conduction band dispersion $\epsilon_k = -2tcosk$ and the symmetric case $E_f = -\frac{U}{2}$, with $U = 5$.

We have investigated the ground-state properties as a function of the hybridization and the energy spacing in the conduction band, $\Delta = 4t/(N-1) = \frac{4t}{5}$. We have calculated the average $f-$ and $c-$local moments, $< (\mu^{f}_{i})^2 >$ and $< (\mu^{c}_{i})^2 >$, and the zero-temperature *f-f* and *f-c* spin correlations functions (SCF) $< S^{f}_{i} S^{f}_{i+1} > \equiv < g|S^{z,f}_{i} S^{z,f}_{i+1}|g >$ and $< S^{f}_{i} S^{c}_{i} > \equiv < g|S^{z,f}_{i} S^{z,c}_{i}|g >$, respectively. Here, $|g >$ is the many-body ground state and $S^{z,f}_{i}$ is the z-component of the f-spin at site i. As expected, the cluster has a singlet ground state ($S_g = 0$ where S_g is the ground-state spin). We compare the onsite Kondo correlation function $< S^{f}_{i} S^{c}_{i} >$ and the nearest-neighbor RKKY correlation function $< S^{f}_{i} S^{f}_{i+1} >$ to assign a state to the Kondo or RKKY regimes, in analogy with mean field treatments[13].

In Fig. 1 we present the variation of the local Kondo SCF $< S^{f}_{i} S^{c}_{i} >$ (squares) and the nearest-neighbor RKKY SCF $< S^{f}_{i} S^{f}_{i+1} >$ (circles) as a function of hybridization for two values of the hopping matrix element $t = 0.2$ (closed symbols) and $t = 1.2$ (open symbols), respectively. As expected, for weak hybridization V the local nearest-neighbor RKKY (Kondo) SCF is large (small), indicating strong short-range antiferromagnetic coupling between the $f - f$ local moments, which leads to long range magnetic ordering for extended systems. As V increases, $< S^{f}_{i} S^{f}_{i+1} >$ decreases whereas the $< S^{f}_{i} S^{c}_{i} >$ increases (in absolute value) saturating at large values of V. This gives rise to the condensation of independent local Kondo singlets at low temperatures, i.e., a disordered spin liquid phase. Interestingly, as t or Δ decreases the *f-c* spin correlation function is dramatically enhanced while the *f-f* correlation function becomes weaker, indicating a transition from the RKKY to the Kondo regime.

In Fig. 2 we present the average local f- (circles) and c- (squares) moments as a function

of hybridization for two values of the hopping matrix element $t = 0.2$ (closed symbols) and $t = 1.2$ (open symbols), respectively. In the weak hybridization limit, the large on-site Coulomb repulsion reduces the double occupancy of the f level and a well-defined local f moment is formed $\langle \mu_f^2 \rangle = 1.0$ while $\langle \mu_c^2 \rangle = 0.5$. With increasing V both charge- and spin-f1uctuations become enhanced and the local $f-$ moment decreases monotonically whereas the $c-$ local moment exhibits a maximum. In the large V limit both the $f-$ and $c-$ local moments have similar behavior with $< \mu_c^2 > \approx \mu_f^2 \approx \frac{1}{2}$, indicating that the *total* local moment μ vanishes. The effect of lowering the energy spacing Δ is to decrease (increase) the $f-$ ($c-$) local moment, thus tuning the magnetic behavior of the system. Note that the maximum value of the $c-$ local moment increases as Δ decreases. This is due to the fact that for smaller t values the kinetic energy of conduction electrons is lowered, allowing conduction electrons to be captured by f electrons to screen the local f moment, thus leading to an enhancement of the local $c-$ moment.

In Fig. 3 we present the energy spacing versus V zero-temperature phase diagram of the nanocluster, which illustrates the interplay between Kondo and RKKY interactions. In the RKKY region $< S_i^f S_{i+1}^f >$ is larger than the $< S_i^f S_i^c >$ and the *total* local moment is non zero; in the Kondo regime $< S_i^f S_{i+1}^f >$ is smaller than the $< S_i^f S_i^c >$, the *total* local moment vanishes, and the ground state of the system is composed of independent local singlets. The red curve indicates the crossover point, i.e., $< S_i^f S_{i+1}^f > = < S_i^f S_i^c >$. The blue dashed curve denotes the set of points where the on-site *total* local moment $\mu = 0$. Thus, in the intermediate regime, which will be referred to as the *free spins* regime[14], $< S_i^f S_{i+1}^f >$ is smaller than the $< S_i^f S_i^c >$, the f moment is *partially* quenched and $\mu \neq 0$. Interestingly, we find that the *free spins* regime becomes narrower as the average level spacing Δ is reduced. This result may be interpreted as a quantum critical regime for the nanochain due to the finite energy spacing, which eventually reduces to a quantum critical point when $\Delta \to 0$.

We have also examined the effect of changing N_{el} from $N_{el} = 12$ ($S_g = 0$) to $N_{el} = 11$ ($S_g = \frac{1}{2}$) for $t = 1$. We find: (a) an enhancement of the local Kondo SCF $< S_i^f S_i^c >$ from -0.01 to -0.12; and (b) a suppression of the f-f SCF $< S_i^f S_{i+1}^f >$ from -0.58 to -0.20 (due to the broken symmetry for $N_{el} = 11$, the f-f SCF's range from -0.5 to +0.02). This interesting novel tuning of the magnetic behavior can be understood in terms of the (single versus double) topmost occupied conduction level: For N_{el} even, double occupancy prevents spin-flip transitions, thus weakening the Kondo correlations.[2]

In conclusion, we have carried exact diagonalization calculations which reveal for the first time that the: (1) energy spacing; and (2) parity of N_{el} give rise to a novel tuning of the magnetic behavior of a *dense* Kondo nanochains. This interesting and important tuning can drive the nanocluster from the Kondo to the RKKY regime, i.e. a tunable Δ verus V zero-temperature phase diagram at the nanoscale. The results indicate the presence of an intermediate *free spins* regime which becomes narrower as the energy spacing is reduced. Our conclusions should be relevant to experimental realizations[4] of small clusters and quantum dots, with appropriate tuning of the energy spacing.

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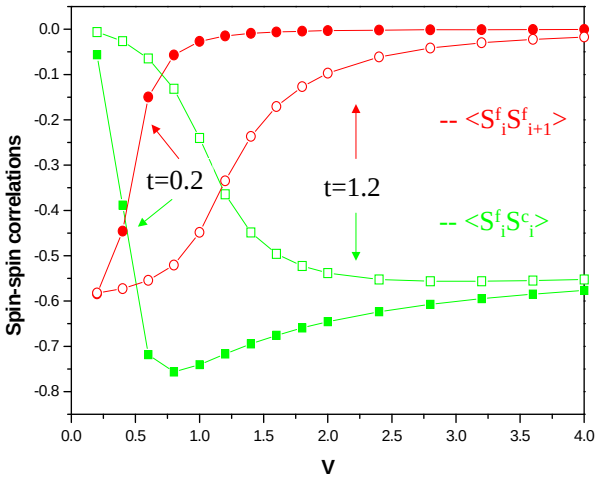

FIG. 1: Nearest neighbor f-f spin-spin correlations (circles) and on-site f-c spin-spin correlations (squares) as a function of V for two values of the hopping parameter of $t = 0.2$ (closed symbols) and $t = 1.2$ (open symbols), respectively.

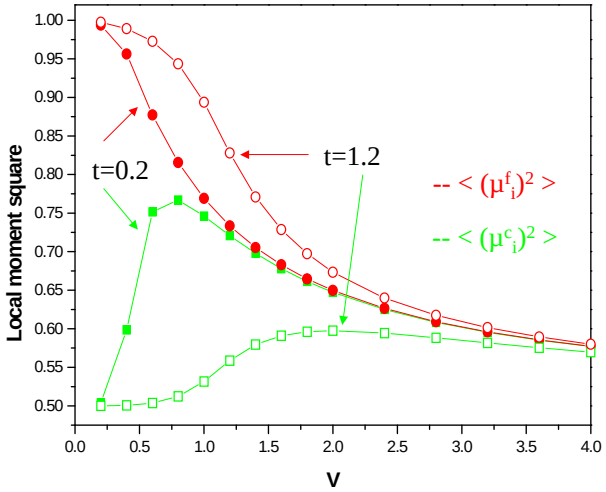

FIG. 2: f- (circles) and $c-$ (squares) local moment versus hybridization for two values of the hopping parameter of t=0.2 (closed symbols) and t=1.2 (open symbols), respectively.

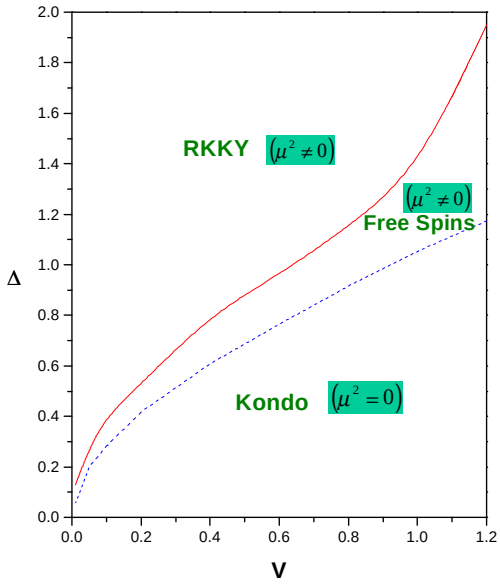

FIG. 3: Energy spacing Δ versus hybridization zero-temperature phase diagram. The red solid curve denotes the crossover point of the spin-spin correlation function in Fig.1; the blue dashed curve denotes the set of points where the on-site *total* moment square $\langle(\mu_f + \mu_c)^2\rangle = 0.0 \pm 0.05$.